



Effectiveness Monitoring for Dioxin Total Maximum Daily Loads in Western Washington

Abstract

Washington State Department of Ecology (Ecology) is required to develop and implement Total Maximum Daily Loads (TMDLs) for impaired waters, and evaluate the effectiveness of the cleanup plan to achieve the needed improvement in water quality. Ecology developed dioxin TMDLs for four mills in Washington State that produce bleached pulp and/or paper products: Weyerhaeuser, Everett; Rayonier, Port Angeles; Simpson, Tacoma; and Weyerhaeuser, Cosmopolis.

The TMDLs were based on the narrative state water quality criterion of 0.014 parts per quadrillion (ppq) at the edge of the mixing or dilution zone. Maximum dioxin target limits of 10 ppq and 5 parts per trillion (ppt) were set for effluent discharge and sludge, respectively, in the NPDES permits to provide protection for human and aquatic organisms as recommended by EPA.

Weyerhaeuser, Everett ceased operation before any discharge monitoring results could be obtained from the facility. Evaluation of available data from the other facilities indicates compliance to the NPDES permit maximum effluent and sludge target limits set for dioxin. Moreover, fish and crab dioxin tissue bioaccumulation results, part of the permit requirements, show the facilities did not appear to pose a problem to public health at the time of data collection.

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Introduction

Over 40% of the nation's and most states' assessed waters still do not meet the water quality standards. Consequently, under the federal Clean Water Act (CWA) section 304(l)(1)(a)(d), the state is required to prepare a list of waters that cannot reasonably be anticipated to attain or maintain water quality standards due to toxic pollutants after the application of effluent limitations required under section 301(b)(2) of the Act. Subsequently, the state was required to develop individual control strategies (ICS) that will result in a reduction in the discharge of toxic pollutants from these point sources, hence the development of these dioxin-related Total Maximum Daily Loads (TMDLs). Washington State Department of Ecology (Ecology) is required to develop and implement TMDLs for watersheds, and evaluate the effectiveness of the cleanup plan to achieve the needed improvement in water quality.

This report presents an assessment of the effectiveness of dioxin-related TMDLs in Western Washington. TMDL is a tool for implementing water quality standards under the CWA and is based on the relationship between pollution sources and in-stream water quality conditions. It is a summation of the individual waste load allocations (WLAs) for point sources and load allocations (LAs) for nonpoint sources, including natural background conditions. Polychlorinated dioxins and furans are not naturally occurring and can be traced to human sources.

Polychlorinated dioxins and furans have about 210 different forms (or congeners) and belong to a class of tricyclic aromatic pollutants that are persistent, toxic and bioaccumulative. The physical/chemical properties of each congener vary according to the degree and position of chlorine and/or bromine substitution. Very little is known about occurrence and toxicity of the mixed (chlorinated and brominated) dioxin, furan, and biphenyl congeners. But much is known about 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD), often simply called dioxin.

The purpose of this report is to assess the effectiveness of past TMDL implementation programs at several Western Washington pulp and paper mills in complying with their National Pollutant Discharge Elimination System (NPDES) permits.

This report includes the following:

- Dioxin background information
- Summary of dioxin NPDES permit limitations in pulp and paper mills' TMDLs
- Discussion on monitoring results from these pulp and paper mills
- Conclusion

Background

Dioxins are produced from: combustion of organic compounds (waste incineration, burning of various fuels, other high temperature sources and poorly controlled combustion sources); metal smelting, refining and processing sources; chemical manufacturing (manufacture of chlorine bleached wood pulp and chlorinated phenols); biological and photochemical processes (microorganisms' activities on chlorinated phenolic compounds); and reservoir sources (potential

re-release from soils, sediments and PCP treated woods) (Alcock and Jones, 1996). Of all these sources, only the chemical manufacturing sources as they relate to pulp and paper mills will be evaluated for this report.

According to Yake et al. (1998), in the EPA's national dioxin source assessment, bleached pulp and paper production was ranked 4th overall as a source of dioxin contamination and may be relatively more important in Washington State. In addition, all the dioxin-related TMDLs in the state are associated with pulp and paper mills.

Currently, there are seven mills in Washington State that produce bleached pulp and/or paper products (Table 1). An eighth one, Rayonier Pulp and Paper Mill in Port Angeles, closed down in 1997 (Yake et al., 1998), while two other mills, Weyerhaeuser Pulp and Paper Mill in Everett, and Georgia-Pacific in Bellingham ceased operation before any monitoring data could be obtained. Hence, no new dioxin was produced at these sites apart from what is left in the environment. Out of all the mills, four TMDLs were developed by Ecology. EPA developed a TMDL for four mills in the Columbia basin and those mills are not part of this analysis. EPA is responsible for TMDL effectiveness monitoring for these mills.

Of the four pulp and paper mills with TMDLs, Discharge Monitoring Reports (DMRs) were obtained from Rayonier Pulp and Paper Mill in Port Angeles before closure and the results are used in this evaluation. There was a shift from elemental chlorine to chlorine dioxide bleaching with Washington pulp and paper mills in the early 1990s, as well as a change in additives used in the paper-bleaching process, which led to less dioxin being produced and released to the environment. Nevertheless, due to the toxicity of dioxins, pulp and paper mills involved in bleaching are required to treat and test their wastewater before discharge. Treated effluent test results for dioxins are sent to Ecology as part of the NPDES permit requirement to evaluate compliance. The maximum dioxin effluent target limit of 10 ppq is used in the NPDES permits as recommended by EPA to provide protection for humans and aquatic organisms.

2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD), otherwise known as dioxin, is the most toxic form of these congeners with four chlorine atoms at specific sites, as indicated in Figure 1.

Dioxins are persistent (resist decomposition), bioaccumulative (accumulate in organisms), and toxic (have deleterious effects on biological organisms). The level of dioxin persistency in environmental media is measured with half life and is the time required for 50% of the compound to degrade through chemical, biochemical and photochemical processes (Environment Canada, 1994). For example, the half life of dioxin in human tissues ranges from 5 to 11.3 years; and 10 to 12 years in anaerobic soils (Atkinson, 1992). Dioxins are relatively insoluble in water, lipophilic in nature and readily accumulate in animal tissues. The rate of accumulation depends on the bioconcentration factor (BCF) which is the ratio of an organism's chemical concentration to that of the organism's surrounding environment, assuming steady-state equilibrium (Environment Canada, 1994). Dioxins in the environment can be ingested by organisms and subsequently transferred through the food chain. Over the years, dioxin accumulation can eventually lead to acute (immediate) or chronic (long-term) toxicity to the organism. The degree of toxicity varies from lethality in juvenile fish to endocrine disruption and reproductive impairment in other organisms.

Table 1. Bleached pulp and paper mill TMDLs and facility status.

Bleached Pulp and Paper Mills	TMDLs Developed	Maximum Dioxin Effluent Detection Limit	Facility Status
Rayonier Pulp and Paper Mill, Port Angeles	Yes	10 ppq	Closed
Weyerhaeuser Pulp and Paper Mill, Longview	Yes, EPA	Set by EPA	Active
Weyerhaeuser Pulp and Paper Mill, Everett	Yes	NA	Closed
Kimberly Clark Pulp and Paper Mill, Everett	No	NA	Active
Weyerhaeuser Pulp and Paper Mill, Cosmopolis	Yes	10 ppq	Active
Simpson Kraft Pulp and Paper, Tacoma	Yes	10 ppq	Active
Fort James Pulp and Paper, Camas	Yes, EPA	Set by EPA	Active
Georgia-Pacific Pulp and Paper, Bellingham	No	NA	Closed
Boise Cascade Pulp and Paper, Wallula	Yes, EPA	Set by EPA	Active
Longview Fiber Pulp and Paper, Longview	Yes, EPA	Set by EPA	Active

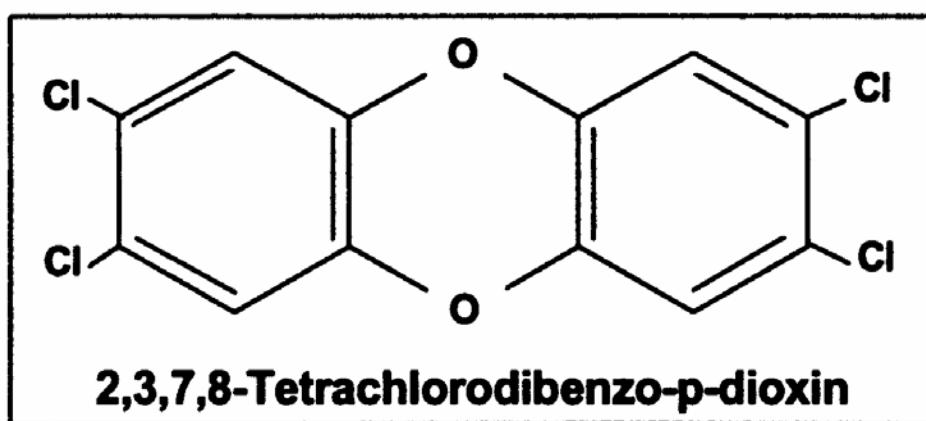


Figure 1. Chemical Structure of 2,3,7,8-TCDD Compound

Pulp and Paper Mills' Dioxin NPDES Permits

Dioxin controls have been established for pulp and paper mills to ensure compliance with the state's narrative water quality standard for toxic substances. A wasteload allocation has been set and implemented through effluent limitations in the NPDES permits. The effluent and sludge target limits are 10 ppq and 5 ppt, respectively (Table 2). The goal for water quality improvement is to keep dioxin loading limits in the final outfall (or discharge) within 0.19, 0.07, 0.19, and 0.18 mg/day for the lower Snohomish River, Commencement Bay, Strait of Juan de Fuca, and Grays Harbor (lower Chehalis River), respectively. These dioxin limits or wasteload allocations are based on three factors: applicable water quality criterion of 0.014 ppq, adequate dilution factor, and background dioxin concentrations. According to the TMDL, this loading rate is consistent with the narrative state water quality criterion of 0.014 parts per quadrillion (ppq) at the edge of the mixing or dilution zone. This criterion is based on a 10^{-6} risk level and ingestion of aquatic organisms; it has no drinking water component. The TMDL also requires that ambient monitoring of the waste stream be conducted by the permittee and that monitoring data be forwarded to the Department of Ecology (see Appendices). Fish and crab tissue bioaccumulation results for dioxin were also collected as part of the permit requirement and the results were used in this analysis.

Table 2. Bleached Pulp and paper Mills' NPDES Effluent and Sludge Limits for Dioxin

No.	Affected Water Body Name	WRIA	TMDL Approval Date	NPDES Effluent Limit (ppq)	NPDES Sludge Limit (ppt)
1	Snohomish River	WA-07	4-Sept-92	10	5
2	Commencement Bay	WA-10	18-June-92	10	5
3	Strait of Juan de Fuca	WA-18	14-July-92	10	5
4	Grays Harbor	WA-22	2-July-92	10	5

Results and Discussion

Effluent

The dioxin results used for this evaluation, with the exception of a few observations, were marked "U". This qualifier denotes the analyte was not detected at or above the reported limit (or less than the given detection limit), as can be seen from the Appendices. No effluent data was available for Weyerhaeuser, Everett (plant closure).

At the Rayonier Pulp and Paper Mill in Port Angeles, the range of dioxin non-detection values in the effluent before the plant closed varied from 0.5 ppq in March 1995 to 10 ppq in January 1993. Most of the measurements were not detected at or above the reported limit. The only real dioxin detection recorded in the history of the data collection was in July 1993 with a value of

5.4 ppq, as shown in Appendix A. All the effluent data, both measured and non-detects, were consistently below the NPDES permit target limit of 10 ppq. In addition, bioaccumulation sample results from locations in the vicinity of the outfall and 5 miles east of the discharge point showed no dioxin in any of the crab tissues sampled. All 1995 crab tissue test results showed non-detectable concentrations of dioxin ranging from 0.11 to 0.18 pg/g, or parts per trillion (ppt) (Rayonier Inc., 1995). Tissue detection limits may have been higher than the standard limit of 0.07 ppt based on the human health water quality criteria contained in 40 CFR 131.36 (known as the National Toxics Rule). These results should be viewed as inconclusive from the standpoint of water quality standard compliance.

The range of dioxin non-detection values in the effluent discharge at the Simpson Kraft Pulp and Paper Mill in Tacoma varied from 0.68 ppq in June 2001 to 8.2 ppq in November 1991, as shown in Appendix B. All readings were non-detectable with no observations exceeding limits. The low concentrations may have resulted from changes in effluent treatment processes by Simpson, specifically, high substitution of chlorine dioxide for chlorine in 1989. According to a Simpson Tacoma Kraft Company Letter in 1994, the company operated dioxin free (non-detect) in its final effluent discharge since 1991, thus reducing any potential for dioxin bioaccumulation. The letter also cited a 1989 EPA crab tissue study and 1991 marine sediment analysis near the Tacoma mill in which the results revealed non-detect levels of dioxin (Simpson Tacoma Kraft Company, 1994). The assumption is that mills with non-detectable effluent dioxin levels contribute little, if any, toward environmental dioxin bioaccumulation. More tissue bioaccumulation data may be required to substantiate this claim.

At the Weyerhaeuser Pulp and Paper Mill in Cosmopolis, the range of dioxin non-detection values in the effluent ranged from 1.0 ppq in December 1995 to 6.0 ppq in August 1994 (Appendix C). Similar to the Simpson Tacoma Kraft Company, the Weyerhaeuser Pulp and Paper Mill in Cosmopolis showed dioxin effluent discharge measurements that were not detected (non-detect) at or above the reported limits. Fish and crab tissue analysis results from sampling locations in the vicinity of the diffuser within the mixing zone, a reference site, a site approximately 1,500 ft east (upstream), and another 1,500 ft west (downstream) in the South Channel were low. A 1994 fish and crab dioxin tissue bioaccumulation study showed that no dioxin was detected in any of the fish and dioxin concentrations in muscle tissues of crabs were very low (CH2M Hill and Weyerhaeuser Paper Company, 1995).

Evaluation of available effluent results indicates compliance with the NPDES permit effluent target limits of 10 ppq for dioxin at all discharging facilities. Beneficial uses of the natural resources, including recreational contact and protection of aquatic life, were protected based on these results. Receiving water impacts appear to be minimal based on the tissue analysis results reported. The assumption is that protection of human and aquatic organisms has been provided in these watersheds barring other sources of dioxin or future accumulation.

Sludge

Based on the TMDL recommendations and the subsequent NPDES permits, the only other source of dioxin (from non combustion sources) from these plants is from sludge. Like the effluent data,

most of the sludge dioxin results used for this evaluation were marked “U”. This qualifier denotes the analyte was not detected at or above the reported limit (or less than the given detection limit). There were no sludge dioxin results available for Weyerhaeuser, Everett and the Rayonier Pulp and Paper Mill in Port Angeles due to plant closure.

The Simpson Kraft Pulp and Paper Mill in Tacoma produced a range of actual dioxin concentrations detected in both primary and secondary sludge products. Actual concentrations in primary and secondary sludge ranged from 0.95 ppt in November 1992 and 0.22 ppt in May 1999 to 2.8 ppt in December 1997 and 1.8 ppt in May 1998, respectively (Appendix B). The rest of the measurements were not detected at or above the reported limit. Figure 2 shows measurable levels of dioxin in sludge to be in compliance with the NPDES permit target limit.

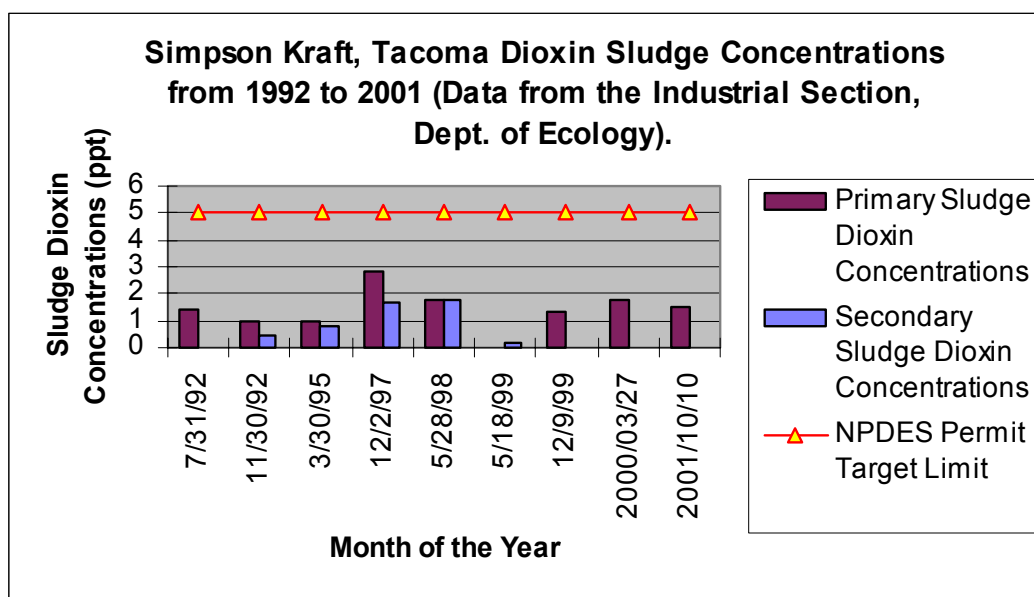


Figure 2. Sludge Dioxin Monitoring Data at the Simpson Kraft Pulp and Paper Mill in Tacoma

The Weyerhaeuser Pulp and Paper Mill in Cosmopolis had a range of dioxin non-detection values in sludge ranging from 0.41 ppt in September 2000 to 19.3 ppt in September 1996 (Appendix C). All sludge readings fell below detection limits, though at times the detection limit was higher than the permit limit. These latter results should be viewed as inconclusive from the standpoint of permit compliance.

Available data suggests general compliance with the NPDES permit sludge target limit of 5 ppt. Compliance is defined as no detection at 5 ppt in sludge samples for these NPDES permits.

Conclusion

Evaluation of available discharge monitoring report (DMR) data and dioxin tissue bioaccumulation results indicates that both effluent and sludge NPDES permit target limits for dioxin are generally being met. The non-detectable concentrations of dioxin in effluent and sludge data and the lack of significant concentration of dioxin in animal tissues collected from the receiving water indicates that progress is being made with TMDL implementation and that compliance with NPDES is the end result. Consequently, these facilities did not appear to pose a problem to public health at the time of data collection. The facilities operations appear to comply with the state's narrative water quality standard for toxic substances; thereby verifying the effectiveness of these four TMDLs. However, future recommendations are to:

- Conduct a current dioxin tissue bioaccumulation study for these mills to verify improvement in water quality, and monitor on a 5-year cycle, and
- Make dioxin receiving water monitoring part of the permit requirement for these mills during future permit renewal.

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Appendix A.

Rayonier, Port Angeles – Dioxin Monitoring Results

Rayonier

NPDES Permit TCDD limit - 0.19 mg/day

Date	Final Effluent		TCDD
	TCDD pg/L	Flow MGD	Discharge mg/day
1993/01/25	< 4.4	37.9	< 0.6
1993/01/26	< 10.0	38.2	< 1.4
1993/01/27	< 7.9	38.6	< 1.2
1993/01/28	< 4.4	37.4	< 0.6
1993/01/29	< 6.6	37.5	< 0.9
1993/06/04	< 9.7	39.8	< 1.5
1993/07/20	5.4	44.0	0.9
1993/10/12	< 5.0	42.8	< 0.8
1994/03/08	< 7.2	37.3	< 1.0
1994/05/30	< 3.8	39.9	< 0.6
1994/08/31	< 2.1	41.7	< 0.3
1994/12/12	< 3.5	40.0	< 0.5
1995/03/21	< 0.5	38.5	< 0.1
1995/05/10	< 4.3	38.4	< 0.6
1995/09/06	< 4.3	33.6	< 0.5
1995/12/20	< 1.5	37.4	< 0.2
1996/03/06	< 2.2	35.0	< 0.3
1996/05/01	< 2.0	36.4	< 0.3
1996/08/06	< 0.98	43.3	< 0.2
1996/11/21	< 4.1	37.6	< 0.6

Mill closed on March 30, 1997

Data from the Industrial Section, Department of Ecology.
 Numbers in **bold** are the real measured dioxin concentrations;
 the rest are detection limits with less than sign meaning non-detect.

Appendix B

Simpson Tacoma Kraft, Tacoma – Dioxin Monitoring Results

Simpson Tacoma Kraft

2,3,7,8-TCDD Data

NPDES Permit TCDD limit - 0.07 mg/day

Date	Secondary Effluent			Primary Sludge	Secondary Sludge
	2,3,7,8-TCDD ppq	Flow MGD	Discharge mg/day	2,3,7,8-TCDD ppt	2,3,7,8-TCDD ppt
1991/11/18	<8.2	27.73	< 0.86		
1992/03/30	<1.7	29.68	< 0.19		
1992/07/31	<3.2	30.64	< 0.37	1.4	<0.25
1992/11/30	<1.9	28.70	< 0.21	0.95	0.41
1993/06/15	<2.5	33.60	< 0.32		
1994/03/30	<2.3	27.00	< 0.24	<0.42	<5.8
1994/06/27	<6.5	29.94	< 0.74	<0.49	<0.37
9/20/94	<1.5	33.72	< 0.19	<0.29	<0.34
1994/12/06	<3.9	27.83	< 0.41	<0.58	<0.66
1995/03/30	<2.3	28.20	< 0.25	1	0.82
1995/06/20	<0.96	30.30	< 0.11	<0.26	<0.32
1995/10/02	<1.1	28.66	< 0.12	<0.54	<0.91
1995/12/11	<6.4	38.36	< 0.93	<0.52	<0.52
1996/03/28	<1.3	25.10	< 0.12	<0.34	<0.36
1996/09/27	<3.2	30.70	< 0.37	<0.42	<0.37
1997/02/06	<2.7	30.62	< 0.31	<0.4	<0.3
1997/06/12	<2.5	30.41	< 0.29	<0.37	<0.31
1997/09/19	<1.9	30.93	< 0.22	<0.45	<0.36
1997/12/02	<0.91	30.59	< 0.11	2.8	1.7
1998/02/24	<2.9	27.30	< 0.30	<0.39	<0.44
1998/05/28	<3.7	26.10	< 0.37	1.8	1.8
1998/11/05	<4.4			<0.59	<0.48
1999/01/27	<1.2			<0.43	<0.21
1999/05/18	<1.9			<0.13	0.22
1999/09/14	<2.6			<3.5	<3.7
1999/12/09	<3.4			1.3	<0.51
2000/03/27	<1.8			1.8	<0.65
2000/06/20	<4.57			<0.99	<0.47
2000/10/06	<1.4			<4.24	<1.12
2001/01/02	<1.11			<1.51	<5.18
2001/03/29	<2.75			<0.903	<0.350
2001/06/28	<0.679				<0.321
2001/10/10	<1.18			1.52	<1.88

Data from the Industrial Section, Department of Ecology.

Numbers in **bold** are the real measured dioxin concentrations;
the rest are detection limits with less than sign meaning non-detect.

Appendix C

Weyerhaeuser, Cosmopolis – Dioxin Monitoring Results

Weyerhaeuser

2,3,7,8-TCDD Data

NPDES Permit TCDD limit - 0.18 mg/day

Date	Pond D Effluent			Pond A Sludge
	TCDD ppq	Flow MGD	TCDD Discharge mg/day	TCDD ppt
1993/03/22	<1	23.6	< 0.09	<0.78
1993/05/03	<3	23.5	< 0.27	<1.08
1993/10/12	<1	26.8	< 0.10	<6.5
1994/12/21	<1	24.8	< 0.09	<1.9
1994/03/22				<13.2
1994/03/23	<2	23.7	< 0.18	
1994/05/24	<2	22.1	< 0.17	<4.1
1994/08/23	<6	25.4	< 0.58	<3.32
1994/11/21				<5.3
1994/11/22	<5	26.2	< 0.50	
1995/01/24	<1	22.0	< 0.08	<5.6
1995/05/30	<2	23.8	< 0.18	<16
1995/08/07	<3	25.5	< 0.29	<7.4
1995/12/12	<1	25.9	< 0.10	<5.03
1996/02/20				<5.1
1996/02/21	<1	22.9	< 0.09	
1996/06/27	<3	23.5	< 0.27	<5
1996/09/24	<3	20.3	< 0.23	<19.3
1996/09/24				<11.5
1996/12/17	<2	27.0		<4.4
1997/04/08				<6.8
1997/04/08				<1.9
1997/01/07	<1	24.9	< 0.09	
1997/01/08				<0.81
1997/05/27	<2	24.7	< 0.19	
1997/07/23				<4.83
1997/07/10	<1	25.0	< 0.09	
1997/10/14	<3	25.9	< 0.29	
1997/10/15				<4.62
1998/04/15				<1.09
1998/01/13	<2	23.4	< 0.18	
1998/01/14				<2
1998/04/15				<1
1998/07/16	<5			
1998/07/20				<4.65
1998/10/06	<4	24.4	0.37	<5.9

1999/01/06				<3.8
1999/01/13	<3			
1999/04/06	<2			<3.7
1999/07/07				<1.2
1999/07/06	<2			
1999/10/05				<1.5
1999/11/09	<2			
2000/01/20				<5
2000/01/25	<2			
2000/04/26	<3			
2000/09/25	<2			<0.41
2000/10/03	<2			<1.01
2001/01/16	<3			<2.61
2001/04/23	<2			<2.43
2001/10/15	<4			<1.08

Data from the Industrial Section, Department of Ecology.